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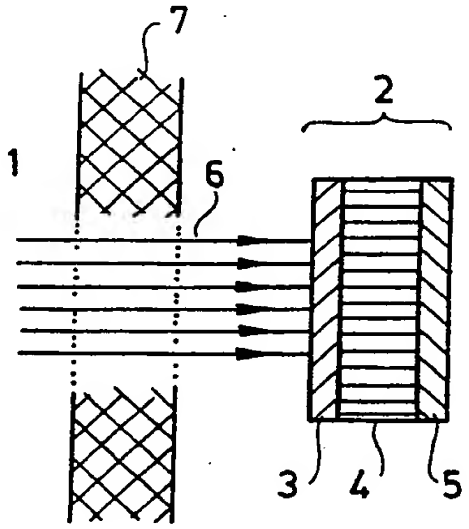
(54) Title: METHOD OF UTILIZING THE (n, gamma) REACTION OF THERMAL NEUTRONS



(57) Abstract

The invention refers to a method of utilizing the (n, gamma) reaction of thermal neutrons, comprising the steps of arranging a target (2) before a source (1) of thermal neutrons, the target (2) having a front surface directed to the source (1) of the thermal neutrons and a rear surface lying behind the front surface, preparing the target (2) with a basic metal body (4) made of $_{70}$Yb and/or $_{74}$W, producing by the means of the thermal neutrons a metallic mixture including the basic metal(s) and at least one of the pairs of metals $_{71}$Lu + $_{72}$Hf and $_{75}$Re + $_{76}$Os and storing the metallic mixture for reducing its nuclear activity.

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METHOD OF UTILIZING THE (n, gamma) REACTION OF THERMAL NEUTRONS

FIELD OF THE INVENTION

The present invention refers to a method of utilizing the (n, gamma) reaction of thermal neutrons, wherein a target is arranged before a source of thermal neutrons. The method of the invention results in possibility of making use of the thermal neutron flux of a nuclear reactor, with disregard to the kind of the reactor, whereby the economy of operating of the different reactors can be highly improved. The proposed method can be realised with reactors of diverse kinds, e.g. with experimental reactors, energetic or boiler reactors etc.

BACKGROUND OF THE INVENTION

It is known from the literature that the (n, gamma) reaction can be applied for producing some isotopes. For example, the reaction

$$^{59}_{27}Co \text{ (n, gamma) } ^{60}_{27}Co$$

is the basis of generating the very important isotope of cobalt having mass numer 60 which is widely used in the medicine and industry. In this process the end product is a substance showing high level of radioactivity (gamma-activity). This process may not be ralised without special security measures.

The theory of atomic nuclei recites lots of theoretical and practical reactions for transforming chemical elements, i.e. atomic nuclei. In the handbooks e.g. the process

$$^{196}_{78}Pt \text{ (n, gamma) } ^{197}_{78}Pt \text{ — } ^{197}_{79}Au$$

can be found for producing gold, wherein the half-period of decay of the intermediate platinum isotope is relatively short, about 20 hours. This way of producing gold is very expensive and inconvenient: the substance at the beginning of the process is twice so expensive than the gold received. Another disadvantage of this process is that the platinum isotope with mass number 196 amounts about 25.3 % of the whole platinum mass and therefore a separate process is necessary for yielding the gold.

SUMMARY OF THE INVENTION

The object of the present invention is to make use of the thermal neutron flux of a reactor for producing non radioactive materials, wherein no special security measures are to be taken.

The invention is based on the recognition that ytterbium and tungsten can be transformed into a mixture of different elements showing no or very low level radioactivity by means of the thermal neutrons generated in each radioactive reactor.

Hence, the invention proposes a method of utilizing the (n, gamma) reaction of thermal neutrons of a reactor, the method comprising the step of arranging a target directed with its front surface to a source of thermal neutrons, especially a reactor, wherein according to the invention the target is consisted of $_{70}Yb$ and/or $_{74}W$. It is especially advantageous to apply before the target a plate shaped body for slowing down the quick and/or the reactor neutrons, consisted of $_{41}Nb$ for slowing down the reactor neutrons and/or $_{59}Pr$ for slowing down the quick neutrons. Of course, this moderator of neutrons can be made also of beryllium. A beryllium plated can be applied also for covering the rear side of the target - this ensures reflection of the neutrons back to the target.

By the means of the method proposed by the invention about 30 % of the amount of ytterbium can be transformed into lutetion and the same amount of tungsten into rhenium. Above that about 20 % of tungsten transform into osmium. The metals received, i.e. lutetium, rhenium and osmium are much more expensive than the input metal of the process and can be separated therefrom by simple thermal processing because of considerable differences in the respective melting points.

BRIEF DESCRIPTION OF THE DRAWINGS

The invention will be further disclosed in more detail by way of example and with reference to the attached drawings. In the drawings

FIG. 1 shows the cross-section of a target applied in realising the present invention.

DETAILLED DESCRIPTION OF THE PREFERRED EMBODIMENTS

In the vicinity of a reactor 1 limited by a wall 7 a target 2 is arranged in an appropriate place. The target 2 consists of a front layer 3 forming a moderating body, a metal plate 4 including ytterbium and/or tungsten to be transformed and a rear reflecting layer 5. The front layer 3 is made of $_{41}Nb$ and/or $_{59}Pr$. If necessary, $_{4}Be$ can be applied to. The mentioned metals slow down the flux of the neutrons leaving the interior of the reactor 2. The reflecting layer 5 covering the rear surface of the metal plate 4 reflects the neutrons back to the metal plate 4. The target 2 is arranged to be irradiated by a thermal neutron flux 6 and the front layer 3 receives the neutrons before entering the metal plate 4.

The neutron flux 6 can be directed to the target 2 through the wall 7 of the reactor 1 in a known way, e.g.

by the means of a window prepared in the wall 7.

As mentioned, the metal plate 4 is made of ytterbium and/or tungsten. The irradiation of this plate carried out by the thermal neutrons generated by the reactor 1 or produced by the front layer in a (n, 2n) reaction should result in an alloy like mixture consisting of the following metals (the composition is given with approximate data):

a) on the basis of ytterbium:

$$\underset{60\ \%}{{}^{37}_{70}Yb} + \underset{30\ \%}{{}^{112}_{71}Lu} + \underset{10\ \%}{{}^{101.4}_{72}Hf} \ (+ \underset{0.1\ \%}{{}^{127}_{69}Tm})$$

b) on the basis of tungsten:

$$\underset{50\ \%}{{}^{19.2}_{74}W} + \underset{30\ \%}{{}^{86}_{75}Re} + \underset{20\ \%}{{}^{15.3}_{76}Os} \ (+ \underset{0.1\ \%}{{}^{21}_{73}Ta})$$

The line over the signs of the elements give the value of the cross-section for the process expressed in barns.

When taking ytterbium, the metal includes the following isotopes:

Mass number of $_{70}Yb$	Proportion	Half-period of decay	Kind of decay
167		19 minutes	gamma, K
168	0.135 %		
169		31.8 days	
170	3.03 %		
171	14.31 %		
172	21.82 %		
173	16.13 %		
174	31.84 %		
175		101 hours	e^-, K
176	12.73 %		
177		1.9 hours	e^-, K

From this table it follows that about 55.3 % of all (n, gamma) reactions do not result in any change of the atomic number. These reactions are:

$$^{170}_{70}Yb \text{ (n, gamma) } ^{171}_{70}Yb$$
$$^{171}_{70}Yb \text{ (n, gamma) } ^{172}_{70}Yb$$
$$^{172}_{70}Yb \text{ (n, gamma) } ^{173}_{70}Yb$$
$$^{173}_{70}Yb \text{ (n, gamma) } ^{174}_{70}Yb$$

The following reactions result in transformation of elements:

$$^{168}_{70}Yb \text{ (n, gamma) } ^{169}_{70}Yb - ^{169}_{69}Tm \quad 100\ \%$$
$$^{174}_{70}Yb \text{ (n, gamma) } ^{175}_{70}Yb - ^{175}_{71}Lu \quad 95\ \%$$
$$^{176}_{70}Yb \text{ (n, gamma) } ^{177}_{70}Yb - ^{177}_{71}Lu \text{ } (e^-, \text{ gamma}) \text{ } ^{177}_{72}Hf \quad 18\ \%$$

The percentage values means the proportion of the given stable isotope in the metal mentioned.

The process of stabilization of $^{177}_{71}Lu$ is characterised by the half-period 6.75 days.

When taking tungsten, the metal includes the following isotopes:

Mass number of $_{74}W$	Proportion, %	Half-period of decay	Kind of decay
179		40 minutes	gamma, K
180	0.135		
181		145 days	gamma, K
182	26.41		
183	14.40		
184	30.645		
185		73.2 days	e^-, gamma
186	28.41		
187		24.0 hours	e^-, gamma

From this table it follows that about 40.8 % of all (n, gamma) reactions do not result in any change of the atomic number. These reactions are:

$$^{182}_{74}W \text{ (n, gamma) } ^{183}_{74}W$$

$$^{183}_{74}W \text{ (n, gamma) } ^{184}_{74}W$$

The following reactions result in transformation of elements:

$$^{180}_{74}W \text{ (n, gamma) } ^{181}_{74}W - ^{181}_{73}Ta \qquad 100\ \%$$

$$^{184}_{74}W \text{ (n, gamma) } ^{185}_{74}W - ^{185}_{75}Re \qquad 37\ \%$$

$$^{186}_{74}W \text{ (n, gamma) } ^{187}_{74}W - ^{187}_{75}Re \qquad 63\ \%$$

In normal circumstances, $^{187}_{75}Re$ is transformed into $^{187}_{76}Os$ characterised by half-period about 5.10^{10} years by weak e^- radiation. In a (n, gamma) reaction, however another process dominates:

$$^{187}_{75}Re \text{ (n, gamma) } ^{188}_{75}Re - ^{188^m}_{76}Os - ^{188}_{76}Os \qquad 13.3\ \%$$

The half-period of rhenium is 18 hours, the isomeric osmium nucleus shows half-period 26 days. In these conditions the rhenium $^{185}_{75}Re$ can be also activated and in decay processes (e^-, gamma, K) it can be transformed partly into tungsten, partly into osmium: a dominate part, however, remains unchanged in form of rhenium.

In both series of reactions the gamma radiation coming into being is a low energy, low intensity weak radiation.

The metallic mixtures prepared by the invention require at least 1/2 year storage before further processing. During this time the radiation level of the mixture falls under a maximum level allowed by the rules.

When considering the basic metal and the metallic components produced bv the method of the invention it can be stated that they are capable of bearing high thermal load and the alloy received in the process is stable. The melting points of the metals in the mixtures mentioned are the following:

$_{70}Yb$	+ $_{71}Lu$	+ $_{72}Hf$	+ $_{69}Tm$	
824 °C	1652 °C	2222 °C	1545 °C	
$_{74}W$	+ $_{75}Re$	+ $_{76}Os$	+ $_{73}Ta$	
3410 °C	3180 °C	2700 °C	2996 °C	

In realising the method of the invention it is advantageous to arrange the target 2 in the proximity of the active zone of the reactor, but under the condition that the target can not be the object of radiation comprising charged particles and fission products. If these factors are excluded the only disturbing effects follow from the gamma radiation of the reactor and the flux of quick neutrons emitted from the reactor. In both cases the loss of neutrons by the nucleus can follow in (gamma, n) and (n, 2n) reactions, however, these are low probability processes. Therefore the only requirement is to moderate the quick neutrons, because the reactions with loss of neutron constitute a part of the reactions which hardly play important rule.

The reactor neutrons show a wide spectrum with average energy 0.72 MeV (the flux may contain also neutrons with energy 20 MeV), therefore it is advantageous to slow down (moderate) the reactor neutrons and the quick neutrons by the means of (n, 2n) reactions whereby the yield of neutrons can be increased. The beryllium moderator is in this case a further element after that applied for slowing down the reactor and quick neutrons.

The reactions of the reactor neutrons are characterized by small cross-section. Hence, they can be slowed down by means of the reaction $^{93}_{41}Nb$ (n, 2n) $^{92}_{41}Nb$. A very effective reaction for slowing down the quick neutrons having energy in the range about 14 to 15 MeV is based on praesodymium: $^{141}_{59}Pr$ (n, 2n) $^{140}_{59}Pr$. The processes mentioned result in increased yield of neutrons. The advantageous character of these reaction can be seen from the following

scheme of reactions:

$$^{41}_{93}Nb\ (n, 2n)\ ^{92}_{41}Nb\ (10\ days,\ decay\ e^-,\ gamma,\ K) -$$
$$-\ ^{92}_{40}Zr\ (n, 2n)\ ^{91}_{40}Zr\ (n, 2n)\ ^{90}_{40}Zr$$

$$^{141}_{59}Pr\ (n, 2n)\ ^{140}_{59}Pr\ (4\ minutes,\ decay\ e^+,\ gamma,\ K) -$$
$$-\ ^{140}_{58}Ce\ (n, 2n)\ ^{139}_{58}Ce\ (140\ days,\ decay\ gamma,\ K) -$$
$$-\ \begin{cases} \ldots\ ^{139}_{57}La \\ (n, 2n)\ ^{138}_{58}Ce \end{cases}$$

Other reaction scheme are possible with low probability, because of the short half-period.

The target 2 includes advantageously a rear reflecting layer 5 for reflecting back the neutrons. This layer can be made of beryllium ($_4Be$).

The plate 4 of the target 2 is arranged preferably so that the neutron flux of the reactor falls under right angle (90°) on its surface.

Summarizing, the method of the invention should be realised with a target 2 including after the reactor a layer made of $^{141}_{59}Pr$ and/or $^{41}_{93}Nb$, a moderator (of $_4Be$), the metal plate 4 made of $_{74}W$ and/or $_{70}Yb$ and a mirror layer (rear reflecting layer 5, made of $_4Be$). The beryllium can be preferred because it is a neutron source under influence of the gamma radiation emitted by the reactor, with the following reactions:

$$^{9}_{4}Be\ (gamma, n)\ ^{8}_{4}Be - 2\alpha$$

wherein the neutrons at the output have energy 110 keV.

The process of the invention can be applied for preparing catalyzer substances – this improves the economy of operating a reactor. No specific security means or expenses are necessary. The metal mixtures can be separated into components according to the known thermal techniques or applied as alloys.

CLAIMS:

1. Method of utilizing the (n, gamma) reaction of thermal neutrons, comprising the step of arranging a target before a source of thermal neutrons, the target having a front surface directed to the source of the thermal neutrons and a rear surface behind the front surface,

characterized in the further steps of

- preparing the target with a basic metal body made of at least one metal selected from the group including $_{70}Yb$ and $_{74}W$,
- producing by the means of thermal neutrons a metallic mixture including the basic metal(s) and at least one pair of metals selected from the group including $_{71}Lu$ + + $_{72}Hf$ and $_{75}Re$ + $_{76}Os$ and
- storing the metallic mixture for reducing its activity.

2. The method as set forth in claim 1, characterized in preparing the basic metal body in the form of a plate and arranging it perpendicularly to the flux of the thermal neutrons.

3. The method as set forth in claim 1 or 2, characterized in the step of arranging on the front surface of the basic metal body a layer for slowing down fast and reactor neutrons by the means of (n, 2n) reactions, the layer consisting of at least one metal selected from the group including $_{41}Nb$ and $_{59}Pr$.

4. The method as set forth in any of claims 1 to 3, characterized in the step of arranging at least one beryllium moderating layer on at least one of the front and rear surfaces of the target.

5. The method as set forth in any of claims 1 to 4, characterized in the further step of carrying out thermal decomposition of the metallic mixture after the storing period.

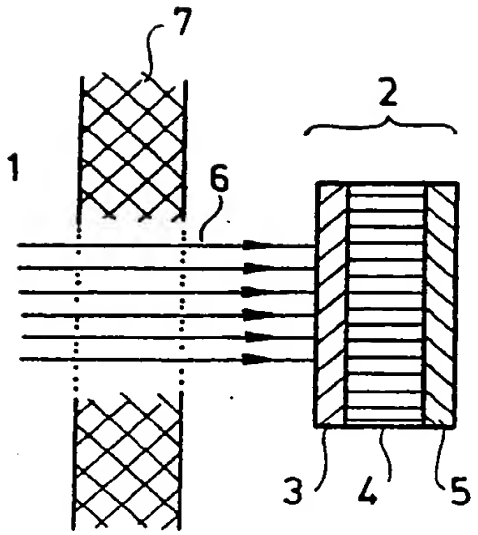

Fig. 1

INTERNATIONAL SEARCH REPORT

International Application No PCT/HU 89/00054

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) [6]

According to International Patent Classification (IPC) or to both National Classification and IPC

IPC[5]: G 21 G 1/06

II. FIELDS SEARCHED

Minimum Documentation Searched [7]

Classification System	Classification Symbols
Int.Cl.[5]:	G 21 G 1/06, 1/08, 4/02; H 05 H 6/00, G 21 K 5/08, G 01 T 1/29

Documentation Searched other than Minimum Documentation to the Extent that such Documents are included in the Fields Searched [8]

III. DOCUMENTS CONSIDERED TO BE RELEVANT [9]

Category [10]	Citation of Document, [11] with indication, where appropriate, of the relevant passages [12]	Relevant to Claim No. [13]
A	GB, A, 1 075 411 (NRA) 12 July 1967 (12.07.67), see claims 7,9.	(1-3)
A	US, A, 4 055 686 (STEINBERG) 25 October 1977 (25.10.77), see claims 2,12; column 6, line 49.	(1-3)
A	GB, A, 974 622 (UNITED KINGDOM ATOMIC ENRGY) 11 November 1964 (11.11.64).	(1-3)
A	DE, A, 1 908 144 (STARK) 11 September 1969 (11.09.69), see claims 2,3.	(1,2,4)
A	DE, A1, 2 941 096 (PHILIPS) 30 April 1980 (30.04.80), see fig. 1b; claims 1,3.	(1-3)
A	GB, A, 440 023 (SZILARD) 12 December 1935 (12.12.35), see page 6, lines 110-130; page 9, line 113 - page 10, line 16.	(1,4,5)
A	GB, A, 1 243 262 (NATIONAL RESEARCH DEVELOPEMENT) 18 August 1971 (18.08.71), see page 2, lines 86, 89; claims 3,8.	(1,2,4)

* Special categories of cited documents: [14]

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IV. CERTIFICATION

Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report
06 February 1990 (06.02.90)	07 February 1990 (07.02.90)
International Searching Authority	**Signature of Authorized Officer**
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III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)

Category *	Citation of Document, with indication, where appropriate, of the relevant passages	Relevant to Claim No
A	W.EPPRECHT, "Werkstoffkunde der Kerntechnik", published 1961, Birkhäuser Verlag, Basel und Stuttgart, see pages 221,323.	(1)
A	K.H.HOCKER, K.WEIMER, "Lexikon der Kern- und Reaktortechnik.", published 1959, Franckh'sche Verlagshandlung W.Keller, Stuttgart, see page 635.	(1)

Anhang zum internationalen Recherchenbericht über die internationale Patentanmeldung Nr.

Annex to the International Search Report on International Patent Application No. PCT/HU 89/00054

Annexe au rapport de recherche internationale relatif à la demande de brevet international n°.

In diesem Anhang sind die Mitglieder der Patentfamilien der im obengenannten internationalen Recherchenbericht angeführten Patentdokumente angegeben. Diese Angaben dienen nur zur Unterrichtung und erfolgen ohne Gewähr.

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Im Recherchenbericht angeführtes Patentdokument Patent document cited in search report Document de brevet cité dans le rapport de recherche	Datum der Veröffentlichung Publication date Date de publication	Mitglied(er) der Patentfamilie Patent family member(s) Membre(s) de la famille de brevets	Datum der Veröffentlichung Publication date Date de publication
GB-A - 1075411		None	
US-A - 4055686	25-10-77	None	
GB-A - 974622		None	
DE-A - 1908144	11-09-69	DE-B2- 1908144	11-08-77
		DE-C3- 1908144	13-04-78
		FR-A5- 2002247	17-10-69
		GB-A - 1243262	18-08-71
		NL-A - 6902545	22-08-69
		NL-B - 162241	15-11-79
		NL-C - 162241	15-04-80
		US-A - 3683190	08-08-72
DE-A1- 2941096	30-04-80	FR-A1- 2438953	09-05-80
		FR-B1- 2438953	30-12-83
		GB-A1- 2033651	21-05-80
		GB-B2- 2033651	18-08-82
		JP-A2-55053899	19-04-80
		NL-A - 7810299	15-04-80
		US-A - 4298804	03-11-81
GB-A - 440023		None	

See page -2-

zu PCT/HU 89/00054

GB-A - 1243262	18-08-71	DE-A - 1908144	11-09-69
		DE-B2- 1908144	11-08-77
		DE-C3- 1908144	13-04-78
		FR-A5- 2002247	17-10-69
		NL-A - 6902545	22-08-69
		NL-B - 162241	15-11-79
		NL-C - 162241	15-04-80
		US-A - 3683190	08-08-72

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